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*J.A. Becker, L.A. Bernstein, W. Younes, D.P. McNabb,
P.E. Garrett, D. Archer, C.A. McGrath, M.A. Stoyer, H.
Chen, W.E. Ormand, R.O. Nelson, M.B. Chadwick, G.D.
Johns, D. Drake, P.G. Young, M. Devlin, N. Fotiades,
W.S. Wilburn*

U.S. Department of Energy

Lawrence
Livermore
National
Laboratory

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Partial γ -Ray Cross Sections for the Reaction $^{239}\text{Pu}(n,2n\gamma_i)$ and the $^{239}\text{Pu}(n,2n)$ cross section

J. A. BECKER¹, L. A. BERNSTEIN¹, W. YOUNES¹, D. P. McNABB¹,
P. E. GARRETT¹, D. ARCHER¹, C. A. McGRATH^{1*}, M. A. Stoyer¹,
H. CHEN¹, W. E. ORMAND¹, R. O. NELSON², M. B. CHADWICK²,
G. D. JOHNS², D. DRAKE^{2†}, P. G. YOUNG², M. DEVLIN²,
N. FOTIADES², W. S. WILBURN²

¹ Lawrence Livermore National Laboratory Livermore, CA 94550

² Los Alamos National Laboratory Los Alamos, NM 87545

Absolute partial γ -ray cross sections for production of discrete γ rays in the $^{239}\text{Pu}(n,2n\gamma_i)^{238}\text{Pu}$ reaction have been measured. The experiments were performed at LANSCE/WNR on the 60R flight line. Reaction γ -rays were measured using the large-scale Compton-suppressed array of Ge detectors, GEANIE. The motivation for this experiment, an overview of the partial γ -ray cross-section measurement, and an introduction to the main experimental issues will be presented. The energy resolution of the Ge detectors allowed identification of reaction γ rays above the background of sample radioactivity and fission γ rays. The use of planar Ge detectors with their reduced sensitivity to neutron interactions and improved line shape was also important to the success of this experiment. Absolute partial γ -ray cross sections are presented for the $6_1^+ \rightarrow 4_1^+$ member of the ground state rotational band in ^{238}Pu , together with miscellaneous other γ -ray partial cross sections. The $n,2n$ reaction cross section shape and magnitude as a function of neutron energy was extracted from these partial cross sections using nuclear modeling (enhanced Hauser-Feshbach) to relate partial γ -ray cross sections to the $n,2n$ cross section. The critical nuclear modeling issue is the ratio of a partial cross section to the reaction channel cross section, and not the prediction of the absolute magnitude.

KEYWORDS: partial γ -ray cross sections, plutonium 238, plutonium 239, reaction cross section

I. Introduction

The absolute partial cross section as a function of incident neutron energy was measured for discrete γ -ray production in the $^{239}\text{Pu}(n,2n\gamma_i)^{238}\text{Pu}$ reaction at LANSCE/WNR. Gamma-ray spectroscopy was accomplished with the large-scale γ -ray array GEANIE. Neutron energy was measured using the time-of-flight technique. The goal was of the experiment was the $^{239}\text{Pu}(n,2n)^{238}\text{Pu}$ cross section, deduced from the measured partial γ -ray cross sections. Partial results from the ^{239}Pu sample measurement are presented here. Measurements were also made on a ^{235}U sample, and these measurements served as a test bed our approach. Complete details of the analysis and complete results including partial γ -ray cross sections for many other gamma rays for ^{239}Pu are presented by Bernstein, *et al.*,^{1,2)} and for ^{235}U by Younes, *et al.*³⁾ Details of the time-of-flight analysis are presented by Younes, *et al.*⁴⁾ The measurement of the absolute efficiency of the large-scale γ -ray spectrometer GEANIE, together with a discussion of the uncertainties relevant for this absolute cross section measurement are described by McNabb, *et al.*^{5,6)} A parallel investigation for the $^{238}\text{U}(n,2n\gamma_i)^{237}\text{U}$ reaction is discussed by Fotiades, *et al.*⁷⁾

The usefulness of the radiochemical ^{238}Pu diagnostic is limited by the imprecise data values of the $^{239}\text{Pu}(n,2n)$ cross section as a function of neutron energy: previously measured values are in poor agreement with each other and also with expectations based on standard nuclear physics. At threshold, the measurements of Mather *et al.*⁸⁾ rise significantly faster

than the measurements of Frehaut *et al.*^{9,10)} At $E_n = 14$ MeV, the measurements of Mather *et al.* and Frehaut *et al.* disagree with each other, and they are in very poor agreement with the precision measurements of Loughheed *et al.*¹¹⁾ It is not surprising that evaluations based on these measurements combined with expectations based on nuclear modeling also disagree with each other.

The techniques of previous measurements of the $^{239}\text{Pu}(n,2n)^{238}\text{Pu}$ cross section fall into 2 categories:

- Direct counting of reaction neutrons (inbeam) produced when the ^{239}Pu sample is bombarded by monoenergetic neutrons. The experimental technique attempts to separate the 2 neutron events of the $n,2n$ reaction from the other reaction channels which also emit neutrons, such as n,n' , $n,3n$, n,f .^{8,9,12)}
- A radiochemical measurement at $E_n = 14$ MeV. Extraordinarily enriched ^{239}Pu was bombarded with a known flux of neutrons. The amount of ^{238}Pu produced was detected by its characteristic α decay, measured offline.¹¹⁾

We bring a new approach to obtaining the $^{239}\text{Pu}(n,2n)$ cross section: a measurement of partial γ -ray cross sections in the reaction $^{239}\text{Pu}(n,2n\gamma)^{238}\text{Pu}$ coupled with an inference from the experimental results of the $n,2n$ cross section (shape and magnitude) using nuclear modeling to relate measured partial γ -ray cross sections to the $n,2n$ reaction channel cross section. The model dependence comes in as the predicted ratio of partial γ -ray cross sections to the $n,2n$ cross section, and not the absolute value of a particular gamma-ray cross section prediction. This is a very important advantage: The predicted

* Corresponding author, Tel. +01-925-422-9676,
Fax. +01-925-422-0883, E-mail: jabecker@llnl.gov

magnitude of an individual partial γ -ray cross section is uncertain because it depends on

- The overall magnitude of the $n, 2n$ cross section, which is the difference between the optical model cross section and the sum of the cross sections for the n, n' , n, xn reactions. The relatively small difference of 2 large numbers, with finite uncertainties, carries a large uncertainty.¹³⁾
- The detailed nuclear structure of the final state nucleus (details of the γ -ray cascade process as the excited ^{238}Pu nucleus decays to the ^{238}Pu ground state).

This new experimental approach is enabled by:

- The use of the Compton suppressed array of Ge detectors (GEANIE) for γ -ray spectroscopy. Ge detectors have excellent energy resolution (FWHM $\sim 1/1000$) and line shape (with suppression) characterized by a peak/total ratio ~ 0.5 at $E_\gamma = 1.3$ MeV. Therefore some ^{238}Pu characteristic γ -rays can be measured over the fission background and target radioactivity.
- The emphasis on planar counters for γ -ray spectroscopy. The ~ 13 mm thick detectors are thick enough to stop the low-energy photon of the ground-state band decay, while background from down scattered γ -rays and scattered neutrons is minimized.
- The intense "white" source of neutrons (pulsed) at LANSCE/WNR¹⁴⁾ together with a well shielded time-of-flight beam line (60R). Neutron flux is measured with a fission chamber. Neutron energy is determined by time-of-flight with respect to the accelerator RF pulse.
- Enhanced reaction models, including direct, preequilibrium, and fission reaction mechanisms, together with Hauser-Feshbach compound nucleus decay.¹⁵⁾

The following sections describe our experimental design, the experimental arrangement and facilities, and a sample of the results with discussion.

II. Experimental Organization

We attacked the issue of developing a reliable $^{239}\text{Pu}(n, 2n)^{238}\text{Pu}$ cross section as a function of E_n in the following way: mount a ^{239}Pu sample in a neutron time-of-flight geometry, irradiate the sample with energetic neutrons produced by the LANSCE "white" neutron source, and measure the characteristic energy of the prompt, inbeam γ -rays produced in the $^{239}\text{Pu}(n, 2n)^{238}\text{Pu}$ reaction as the excited nucleus ^{238}Pu decays. The neutron energy (for a given event) is obtained by time-of-flight, and the characteristic gamma-rays signal the decay of a particular level in ^{238}Pu . Analysis proceeds via construction of 2-D data matrices with neutron energy E_n and E_γ as the x- and y-axes, respectively, and number of counts as the z-axis. Gamma-ray yields as a function of neutron time-of-flight are obtained by stepping through energy cuts on the neutron time-of-flight axis, with analysis of the γ -ray spectra to obtain characteristic γ -ray

yields at each cut. Identify discrete γ -rays in the spectrum with the γ -ray transition in the residual nucleus (^{239}Pu , ^{238}Pu , fission fragment, sample activity). Convert time-of-flight to incident neutron energy, correct the appropriate gamma-ray yields for γ -ray spectrometer efficiency, angular distribution, internal conversion, incorporate neutron flux, and the result is a partial γ -ray cross section. The final step is to infer the reaction cross-section shape and magnitude as a function of neutron energy using nuclear modeling to relate partial cross section to reaction cross section.

1. Experimental Arrangement

The sample (^{239}Pu) is placed at the focal volume of GEANIE, a large-scale array of suppressed Ge detectors. GEANIE is located on the LANSCE/WNR 60R neutron flight line, 20.34 m from the spallation neutron source. The spallation neutrons are produced by the interaction of a pulsed 800 MeV proton beam delivered by the LANSCE accelerator onto a W rod. The proton beam average current in 1998 was 2 μA , and in 1999 it was 6 μA . A typical pulse structure for these experiments was: 100 Hz macro pulses, with each macro pulse consisting of a pulse train with a mark-space interval of 1.8 μs and duration 625 μs . The time-of-flight for a 1 MeV neutron is $t = 1.47$ μs . "Wrap around" neutrons (spill over neutrons at the sample from adjacent micro pulses) happens at $E_n = 0.6$ MeV, and therefore events due to "wrap around" neutrons are not an issue for the $^{239}\text{Pu}(n, 2n)$ reaction with Q value -5.64 MeV. Neutron flux at the target was monitored using a fission chamber containing separate ^{238}U and ^{235}U foils and located just upstream of GEANIE in the neutron flight line. Fission events were counted, and neutron flux deduced using the "standard" fission cross sections. Events from adjacent pulses are an issue for the ^{235}U fission foil, but not the ^{238}U fission foil, because of the different (n,f) reaction thresholds. The actinide sample was larger in diameter than the neutron beam spot size on the sample. Absorbers of Pb and Polyethylene in the flight line reduced the γ -flash (produced when the proton beam pulse strikes the W target) and hardened the neutron flux. Charged particles produced in the spallation reaction were prevented from reaching the sample by magnetic deflection.

2. Signal processing and data acquisition

Signal processing was accomplished with standard electronic modules of nuclear science. Data acquisition was done on an event-by-event basis. A γ -ray hit in a Ge counter produced both a linear signal proportional to γ -ray energy deposition in the Ge counter, and a relative time signal relative corresponding to neutron production at the W spallation target. The event stream (gamma-ray pulse height, fission pulse height, relative event time, detector channel ID) was stored onto magnetic tape for offline analysis, and sampled online. High-speed scalars were recorded periodically, merged into the data stream, and therefore also stored onto tape throughout the data acquisition period. This was particularly useful to monitor individual detector dead time. In 1999 data was also tagged with a beam/on – beam/off register and a clock time re-

set every macropulse. An important feature of the data acquisition system was that all data channels were treated equally. Signal processing channels following the preamplifiers were identical in hardware for both the Ge detectors and the fission chamber used to monitor neutron flux; timing signals from these channels were mixed to provide the event trigger, and thus all data channels see the same data acquisition system dead time. Therefore the ratio of γ -ray events to neutron flux is exceptionally stable, with corrections for count rate fluctuations in different detector channels not important to first order.

Data acquisition was accomplished with the computer based data acquisition developed at the National Superconducting Cyclotron Laboratory: The 4π Data Acquisition system.^{16,17} Local customization of data acquisition system for these experiments is described by Younes *et al.*¹⁸ Data acquisition intervals for a given sample amounted to weeks, and beaming periods were months.

3. Data reduction and analysis

The data reduction and analysis for these two experiments was done at LLNL. The basic idea of the analysis is to sort the event stream (gamma-ray pulse height, neutron flight time, and ID) into 2-D matrices of time vs. γ -ray pulse height. All γ -ray channels were added together after energy alignment without loss of energy resolution. (All linear amplifiers ADC's were identical and showed the same non-linearity). Cuts are made in time-of-flight and spectra (histograms) of γ -ray pulse height extracted. Analysis of the peaks in the γ -ray pulse height spectra produces the γ -ray yield corresponding to a particular time-of-flight cut. The desired end product is gamma-ray yield as a function of incident neutron energy E_n ; therefore time-of-flight has to be converted to E_n . This treatment is complicated by the time response function of the Ge detectors to an event. This time response is approximately Gaussian, with FWHM $\sim 10 - 20$ ns. The time response also depends on the energy deposition in the detector. Therefore a time-of-flight cut is not sharp in neutron energy but contains contributions from the adjacent time bins due to the time resolution of the Ge detector, and it is important to account for this time dispersion in cross section. Younes *et al.*,^{3,18} has described our approach to this issue and the analysis protocol developed. The protocol was used at LLNL in the analysis of the ^{239}Pu sample.

4. Samples and sample preparation

The ^{239}Pu sample used in this experiment is of special note because of its (1) high enrichment in ^{239}Pu and (2) exceptionally low ^{241}Am content (≈ 1 ppm). The ^{239}Pu sample was obtained from material reprocessed especially for this experiment in 1999, beginning with a molten salt reduction. After standard reprocessing, the material was heated and levitated in vacuum to drive off the Am,¹⁹ taking advantage of the different vapor pressures of Pu and Am. Two ^{239}Pu samples (as discs) were prepared from this material with nominal thickness of 10 and 20 mils, and diameter 1.25 inches. The samples were surrounded by a Monel ring, and double encapsulated between 2 Be foils (10 mil thick) on each side of the

ring. Carbon foils (5 mg/cm^2) inserted between the inner Be foil and the Pu disk ranged out the α particles associated with the Pu decay, and thus eliminating a background source of γ radiation, the $^9\text{Be}(\alpha, n)^{12}\text{C}$ reaction.

5. Mixed sample irradiation

The 1999 data set had an additional feature: the irradiated sample was a layered composite of $^{239}\text{Pu} + \text{Fe}$. The precision energy resolution and line-shape characteristics of the GEANIE detectors enables separation of the ^{56}Fe 847-keV γ -ray from the neutron induced γ -rays on the actinide samples, and also the radioactivity of the actinide samples. The $^{56}\text{Fe}(n, n'\gamma_{847})^{56}\text{Fe}$ is a standard cross section at 14 MeV and therefore we have an overall check on the entire experiment.²⁰ The Fe foils were natural Fe, 0.2 mm thick, and 2 each were placed on either side of the ^{239}Pu encapsulations, for a total of 4 foils. The advantage of this "mixed sample" technique is that γ -rays produced by neutrons interacting with the Fe sample and the actinide sample are counted simultaneously and processed together. The GEANIE spectrometer does not distinguish a Fe γ -ray from an actinide γ -ray, apart from energy deposition in the Ge detectors. Verification of the $^{56}\text{Fe}(n, n'\gamma_{847})^{56}\text{Fe}$ cross section at $E_n = 14$ MeV means that our understanding of spectrometer response (efficiency, dead time...) and our derivation of the neutron flux and counting corrections is properly done, at least at $E_n = 14$ MeV.

In the worst case, we obtain ratios of the ^{239}Pu to ^{235}U cross sections, using the Fe 856-keV gamma-ray yields as a neutron flux monitor. In practice, the Fe cross-section standard was reproduced within 1 standard deviation.

6. Neutron Beam Spot and GEANIE Efficiency

The absolute cross section measurement requires not only sample characterization, but also measurements of the neutron flux as a function of neutron energy, and finally the efficiency of the GEANIE spectrometer for γ -ray detection in real time. The efficiency is complicated because it depends on:

- The response function of the Ge detector to incident γ -rays (which is a function of E_γ)
- The fluctuating instantaneous counting rate (which produces rate dependent dead time); detector dead-time is also dependent on detector position in the array
- The size and distribution of the neutron beam spot on the sample
- Gamma-ray absorption in the Pb collimators of the individual detector channels, self-absorption in the actinide sample, and absorption in the encapsulation

The γ -ray efficiency of the GEANIE spectrometer was mapped with radioactive sources (calibrated and traceable to a NIST standard). The "point" radioactive sources were moved in space over the area of the neutron beam spot. The beam spot itself was mapped using image plates. (The roughly circular beam spot (~ 2 cm diameter), was smaller than the Pu or U

samples to ensure every neutron passing through the flux monitor strikes a well characterized sample.) The GEANIE spectrometer, beam spot, sample, and encapsulation were modeled in MCNP. The Monte Carlo calculations and measurements compared favorably, and the Monte Carlo calculations were used to extrapolate efficiencies to γ -ray energies between the measured energies of the point sources.⁵⁾ Finally, counting losses due to pulse pile-up (also a function of detector channel) were calculated from a comparison of the summed counts in the individual γ -ray spectra and the corresponding scalar reading. Scalar dead-time effects are roughly a factor of 20 \times less than pileup in the energy spectroscopy channel.

III. $^{239}\text{Pu}(n,2n)^{238}\text{Pu}$ Partial Gamma-ray Cross Sections

The lowest member of the ground state band that could be identified is the $^{238}\text{Pu } 6_1^+ \rightarrow 4_1^+$ transition. The $^{238}\text{Pu } 4_1^+ \rightarrow 2_1^+$ γ -ray was overwhelmed by a fission product γ -ray, and the decay of the $^{238}\text{Pu } 2_1^+$ level is so highly converted that no attempt was made to include it in the experimental design. Identification of the spectral peak with the $^{238}\text{Pu } 6_1^+ \rightarrow 4_1^+$ transition was made on the basis of its well known γ -ray energy, $E_\gamma = 157.4$ keV, and its behavior with incident neutron energy. A standard peak-fitting procedure was applied, and γ -ray yields were extracted, time-cut by time-cut, from the 2-D data matrix of γ -ray pulse height vs. time. The peak yields were corrected for γ -ray efficiency, angular distribution, counting losses, internal conversion, neutron flux and sample characterization to produce an absolute cross section for the $^{238}\text{Pu } 6_1^+ \rightarrow 4_1^+$ transition, which is illustrated in **Figure 1**: The experimental cross section shows the appropriate trends, rising near threshold, peaking, and decreasing with the opening of the $n,3n$ channel. The shape of this partial cross section is well predicted by the model calculations. The absolute magnitude is, in general, not well predicted since it depends strongly on model input choices which determine the overall magnitude of the $n,2n$ channel, and nuclear structure details not in the model which determine the details of the γ -ray cascade at low-excitation energy. Consequently, not just the magnitude, but also the ratio of γ -ray yields may also not be well predicted. In the present case, there is another strong ^{238}Pu γ -ray in the spectrum, the (936.6, $4_1^- \rightarrow 4_1^+$) transition. This transition represents the decay of the bandhead of the $K = 4, J^\pi = 4^-$ quasiparticle band at $E_x = 1083$ keV. It has approximately 1/2 the cross section of the $^{238}\text{Pu } 6_1^+ \rightarrow 4_1^+$ transition at $E_n = 10$ MeV, more than expected. This quasiparticle band apparently acts as a K trap in the γ -ray cascade path. Model predictions are improved, in practise, by including the experimental partial-level scheme data in the modeling of the γ -ray cascade, but the required experimental information does not exist to high enough excitation in ^{238}Pu .

IV. $^{239}\text{Pu}(n,2n)^{238}\text{Pu}$ Reaction Cross Section

Reliable estimates of the reaction cross section can be made when the $2_1^+ \rightarrow 0_1^+$ partial γ -ray cross section is measured, since it carries about 90% of the cross section. Here, the low-

est γ ray measured in the ground state band is the 157.4 keV, $6_1^+ \rightarrow 4_1^+$ transition. The discussion above suggests that a reliable value of the $n,2n$ reaction cross section cannot be estimated on the basis of any single partial cross section measured in this experiment. However, the appropriate sum of all the independent γ -ray paths must be equal to the $n,2n$ cross section, which suggests the procedure adopted here: average over the γ -ray transitions by summing the measured partial cross sections for all the independent paths of the γ -ray cascade (near yrast, at low-excitation energy), and compare with the same ratio predicted by nuclear modeling. This is expressed as:

$$\sigma(n, 2n) = \sum_{\gamma_i} \sigma^{exp}(n, 2n\gamma_i) \times \frac{\sigma^{model}(n, 2n)}{\sum_{\gamma_i} \sigma^{model}(n, 2n\gamma_i)} \quad (1)$$

The value of the $^{239}\text{Pu}(n,2n)^{238}\text{Pu}$ reaction deduced from the experimental results²⁾ and model calculation results²¹⁾ with Eq. (1) is illustrated in **Figure 2**, together with evaluations for comparison.²²⁻²⁵⁾ Input data included the partial cross sections for the 5 strongest ^{238}Pu γ -rays observed. The partial γ -ray cross sections included are (E_γ in keV, assignment): (157.4, $6_1^+ \rightarrow 4_1^+$), (936.6, $4_1^- \rightarrow 4_1^+$), (918.7, $1_1^- \rightarrow 2_1^+$) (corrected for branching²⁾), (924.0, $2_1^- \rightarrow 2_1^+$), and (617.3, $5^- \rightarrow 6^+ / 3^- \rightarrow 4^+$ doublet). The deduced cross section has the overall shape as a function of E_n expected from nuclear modeling, rising smoothly near threshold, flat near $E_n = 11$ MeV, and decreasing as the $n,3n$ channel becomes energetically allowed. There is some uncertainty at the lowest values of E_n near threshold, where the cross section appears to be somewhat high, and there is some uncertainty above $E_n = 12$ MeV, where the modeling of preequilibrium and associated angular momentum issues becomes uncertain. The cross section reported here agrees with the recent value deduced on the basis of systematics of actinide cross sections by Navratil, *et al.*,²⁶⁾ at $E_n = 11xxx$ MeV, and it agrees with the radiochemical value deduced by Loughheed *et al.* for $E_n = 14xxx$ MeV, as evaluated by McNabb and Chadwick.²⁷⁾ This cross section is not in good agreement with any of the previous evaluations. Finally, the particular model calculation used in Eq. (1) included an ^{238}Pu partial level scheme extended by 15 additional levels based on systematics of ^{236}U and ^{240}Pu , developed by Chen.²⁸⁾ Chen, *et al.* also describe an independent model calculation. The cross section results are reasonably independent of level scheme (extended or not) and particular model.

V. Conclusion

The absolute shape and cross section for the $^{239}\text{Pu}(n,2n)^{238}\text{Pu}$ reaction have been incorporated into a new cross-section evaluation,²⁷⁾ significantly improved over past evaluations. The end result is greater confidence in interpretation of a radiochemical diagnostic that contributes to an understanding of the magnitude and energy dependence of a neutron fluence spectrum, and which is therefore important to the Stockpile Stewardship program.

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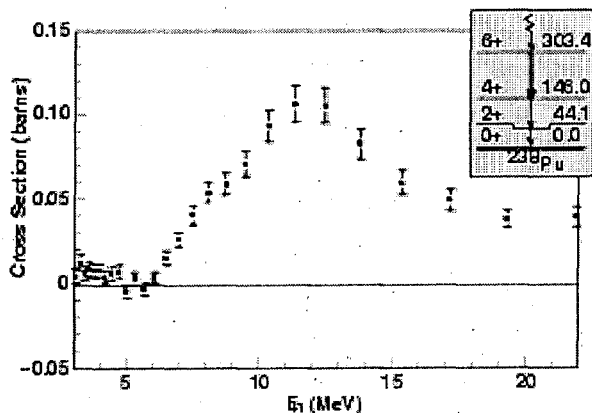


Fig. 1 The partial γ -ray cross section for production of the $6^+ \rightarrow 4^+$ transition, $E_\gamma = 157.4$ keV, in the $^{239}\text{Pu}(n, n)^{239}\text{Pu}(E_x = 303 \text{ keV})$ reaction. The ordinate errors include both systematic and statistical contributions, approximately equal.

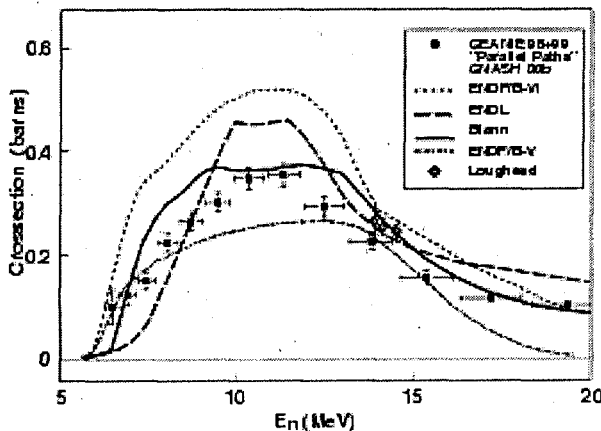


Fig. 2 The $^{239}\text{Pu}(n, n)$ cross section deduced from the present measurement using the reaction code predictions. Evaluations of the cross section, along with the revised evaluation of the results of Loughheed, *et al.* are included for comparison. References to the evaluations are given in the text.

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